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EUROPEAN PATENT APPLICATION

⑲ Application number: 88120630.4

⑤① Int. Cl.4: **C08L 23/02 , C08L 23/16 ,**
C08L 25/04

⑳ Date of filing: 09.12.88

③① Priority: 11.12.87 IT 2294687

④③ Date of publication of application:
14.06.89 Bulletin 89/24

⑧④ Designated Contracting States:
AT BE CH DE ES FR GB IT LI NL SE

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⑤④ Elastoplastic compositions and process for preparing them.

⑤⑦ Elastoplastic compositions comprising a continuous crystalline polyolefin phase and at least two discrete phases dispersed in the polyolefin phase, of which one consists of a dynamically cured EPDM rubber while the other of an amorphous and thermoplastic styrene polymer, in which at least 80% of the dispersed particles has a maximum size preferably below 5 μ m.

The compositions are prepared by masticating the component mixtures, under rubber dynamic curing conditions.

EP 0 320 001 A2

Elastoplastic Compositions and Process for Preparing them

The present invention relates to elastoplastic compositions comprising a continuous crystalline polyolefin phase and at least two polymeric phases dispersed in the polyolefin matrix, of which one phase consists of a dynamically cured EPDM rubber and the other phase consists of an amorphous and thermo plastic styrene polymer.

5 Elastoplastic compositions based on crystalline polymers of olefins and of dynamically cured EPDM rubbers are well known in literature.

Such compositions and the preparation thereof by mixing the components under rubber dynamic curing conditions are described in particular in U.S. patents 3,806,558, 4,130,535 and 4,311,628.

10 The compositions prepared by the dynamic curing method exhibit the drawback, which is common to all the compositions based on plastomeric resins and on cross-linked elastomers, of being the more difficult to be processed, the higher is the percentage of cross-linked elastomeric component contained in them.

For example, compositions comprising a cross-linked EPDM rubber in an amount higher than 70-75 % by weight referred to the polyolefin phase are fully unprocessable.

15 The most interesting curative systems utilized for the rubber curing exhibit furthermore the drawback of giving rise to remarkable corrosion phenomena of the equipment during mastication of the components.

Thus, there is a growing necessity to have available elastoplastic compositions which are readily processable and do not require the use of curative systems leading to the abovesaid corrosion phenomena.

20 It was suggested, in the case of compositions comprising a crystalline olefinic polymer and a uncured saturated ethylene-propylene rubber, to incorporate styrene polymers into said compositions in order to improve the elastomeric characteristics of the compositions (published Japanese patent application No. 17137 83).

The compositions disclosed in the Japanese application are prepared by hot mixing the polyolefin and the rubber, by operating in the presence of styrene and of a peroxide.

25 In these compositions the rubber is uncured, wherefore the processability problems, which are typical of the compositions in which the rubber is present in the cured state, are not encountered.

30 It has now surprisingly been found that it is possible to obtain elastoplastic compositions comprising a crystalline olefin polymer and a cured EPDM rubber, endowed with improved processability characteristics even in the presence of a high content of cured rubber, and which do not require the use of curative systems leading to corrosion phenomena, when the polyolefin phase comprises, besides the cured rubber, also a thermoplastic and amorphous styrene polymer in the form of dispersed particles, of which at least 80% has a maximum size preferably below 5 μm .

35 Thus, the compositions of the invention comprise a continuous crystalline polyolefin phase and at least two polymeric phases dispersed in the polyolefin phase, one of which consists of a dynamically cured EPDM rubber and the other of an amorphous thermoplastic styrene polymer, wherein the EPDM rubber is a copolymer of propylene with ethylene and/or with an alpha olefin of formula $\text{CH}_2 = \text{CHR}$ in which R is an alkyl radical having 2-10 carbon atoms, and with a copolymerizable diene, the weight ratio between polyolefin resin and EPDM rubber ranges from about 10/90 to about 75/25 and the weight ratio between styrene polyester and polyolefin ranges from about 10/90 to about 60/40. Advantageously in the compositions according to the present invention at least 80% of the particles of the dispersed EPDM rubber have a 40 maximum particle size below 5 μm .

Preferably, the particle sizes of both the dispersed phases are such that at least 80% has a maximum size below 5 μm particularly below 2 μm .

Determination of the particle size is carried out by electronic microscopy.

45 Utilizable styrene polymers are all the amorphous polymers and copolymers having thermoplastic nature.

Examples of such polymers are "crystal" polystyrene (PS), impact polystyrene (HIPS), the thermoplastic copolymers of styrene with acrylonitrile and maleic anhydride, the partially hydrogenated styrene-butadiene block copolymers.

50 Examples of partially hydrogenated styrene-butadiene block copolymers are represented by the copolymers described in U.S. patent 4,107,130.

Polystyrene (PS and HIPS), the copolymers of styrene with acrylonitrile containing up to 30% by weight of acrylonitrile, the styrene-ethylene-butene block copolymers (produced by Shell Oil and known under the trade name KRATON® G 1652) are the preferred materials.

The compositions according to the present invention are prepared by means of a process in which the olefin polymer and the styrene polymer, homogeneously mixed with each other, are added and mixed

with EPDM rubber and the curative system is added to the resulting homogeneous blend, mastication of the blend being continued at a temperature between about 150° and 280° C for a time period in which rubber curing occurs.

In order to obtain dispersions, in which the sizes of the particles have the above-indicated values, it is necessary to utilize, during the homogenization step of the styrene polymer with the polyolefin resins, suitable compatibilizing agents such as graft copolymers of styrene on polyolefins, graft copolymers of EPDM rubber on polystyrene or on styrene-acrylonitrile copolymers and the styrene-propylene block copolymers.

These compatibilizing agents are utilized in an amount generally ranging from 5 to 50% by weight calculated on the polyolefin-polystyrene blend.

Styrene block copolymers containing blocks of monomeric units compatible with the polyolefins, such as the styrene-ethylene-butene block copolymers, are also suited to prepare dispersions having the desired dimensional characteristics.

With a view to obtaining a homogeneous dispersion of the polystyrene phase it is advisable to previously prepare an alloy with the polyolefin; such alloy is then utilized to prepare the elastoplastic compositions of the invention.

For the preparation of the alloys, all the methods suitable for obtaining an intimate mixing and homogenization of the components are utilizable. For example, it is possible to operate in an internal mixer or in an extruder or in a system composed of a mixer and a granulator.

For example, an alloy can be properly prepared by dry-mixing the polyolefin in a turbomixer, and optionally HIPS polystyrene, in the presence of a peroxide and of styrene added in such amount as to form the desired percentage of polystyrene homopolymer and of graft copolymer of styrene on the polyolefin. It is operated at temperatures at which no softening and consequent thickening of the polymer can occur.

As regards the sizes of the dispersed particles of the styrene polymer present in the alloys, it has been found that final compositions can be obtained still having satisfactory properties even when the dispersed phase or at least 80% of it has a maximum size of 40 μm .

The useful crystalline polyolefin resins comprise (high, mean or low density) polyethylene and the polymers of the alpha olefins of formula $\text{CH}_2 = \text{CHR}$ in which R is an alkyl radical of 1-8 carbon atoms prepared by using Ziegler-Matta stereospecific catalysts.

High isotacticity index polypropylene is the preferred polymer. Further useful alpha polyolefins are polybutene, poly-4-methyl-1-pentene, polyhexene.

In the compositions of the invention the olefin polymer can be present in a modified form as compared with the starting polymer. That is due to interactions with the curative system, the styrene polymer and the EPDM rubber, which can cause also a sensible lowering of the crystalline melting point (determined by D.S.C.).

The EPDM rubbers are copolymers of propylene with ethylene and/or with another alpha olefin having formula $\text{CH}_2 = \text{CHR}$ in which R is an alkyl radical of 2-10 carbon atoms, and with a diene monomer which is present preferably in an amount ranging from 1 to 10% by weight calculated on the copolymer total weight. Preferably the diene is of the non-conjugated type.

Suitable diene monomers are for example 1,4-hexadiene; 2-methyl-1,4-pentadiene; 1,4,9-decatriene; 1,5-cyclooctadiene; 1-methyl-1,5-cyclooctadiene; 1,4-cyclopentadiene; dicyclopentadiene; ethylidenenorbornene; 4-methyl-1,4-hexadiene; 5-methyl-1,4-hexadiene; the substitution derivatives of such monomers.

Example of olefin monomers of formula $\text{CH}_2 = \text{CHR}$ are propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 3-methyl-1-pentene, 3,3-dimethyl-1-butene, 3-methyl-1-hexene, 2,4,4-trimethyl-1-pentene.

The ethylene-propylene-diene terpolymers containing from 25 to 50% by weight of copolymerized propylene units are preferred.

In the compositions, the olefin polymer/EPDM rubber ratio is in the range of from about 10/90 to about 75/25, e.g. from about 10/90 to about 60/40 and preferably from 15/85 to 50/50; the styrene polymer/polyolefin ratio is generally in the range of from about 10/90 to about 60/40 and preferably from 30/70 to 50/50.

The ratio between the total weight of the polyolefin and of the styrene polymer and the weight of the EPDM rubber generally ranges from 20/80 to 70/30 and preferably from 25/75 to 60/40.

Mineral fillers, carbon black, colored pigments, plasticizers, stabilizers, extender oils, and in general all the conventional ingredients of the elastoplastic compositions comprising EPDM rubbers can be present in the compositions of the invention.

The compositions are prepared by masticating homogeneous blends of the components, under EPDM rubber dynamic curing conditions.

It is possible to operate according to the dynamic curing methods described in U.S. patents 4,130,535

and 4,311,628, utilizing the curative systems therein described.

However it has been found - this being an additional feature of the invention - that it is not necessary, for preparing the compositions of the invention, to use curative systems of corrosive nature like those comprising a phenolic resin and an activator.

5 A fully unexpected result resides in that it is sufficient to use a non-halogenated phenolic resin alone without using activators such as hydrated tin salts and organic acids such as the oxalic, salicyclic, malonic and succinic acids.

Non-halogenated phenolic resins are described in U.S. patents 3,287,440, 3,709,840 and 4,311,628.

Useful non-halogenated resins are also available on the market; for example, such resins can be 10 purchased from Schenectady Chemicals Inc. under the trade name FXRA-148.

The temperature conditions under which mastication is conducted (ranging from 150° to 280° C) and the shear rate employed (300-400 s⁻¹) are furthermore sensibly lower than the ones utilized so far.

According to a preferred procedure, a homogeneous blend of olefin polymer, styrene polymer with the EPDM rubber and, optionally, with fillers is prepared by operating at a temperature sufficient to melt the 15 olefin polymer and for a time sufficient to obtain a homogeneous blend. The phenolic resin is then added and mastication is continued at a temperature at which rubber cure occurs.

Preferably, the EPDM rubber is fully cured. Full curing of the rubber means the cure in which the rubber is extractable for less than 2% with cyclohexane at room temperature or for less than 4% with boiling xylene (as regards the methods of determining the extractability in cyclohexane and xylene, 20 reference is made to U.S. patent 4,806,558).

The blending and/or mastication process can be carried out in an internal mixer, or in an extruder, or in a system consisting of an internal mixer and of a granulator.

It is possible also to operate in a plurality of apparatuses arranged in series, in the first ones the intimate blending and homogenization of the composition occurs while curing takes place in the others.

25 The mastication temperature at which curing occurs is generally in the range from 150° to 280° C and preferably from 180° to 220° C.

The following examples are given merely to illustrate the invention and are not to be considered as a limitation of the scope thereof.

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Example 1

Table 1 shows the composition of a reactive mixture utilized for the preparation of polypropylene-polystyrene alloys useful to prepare the elastoplastic compositions of the invention.

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Table 1

COMPOSITION OF THE REACTIVE MIXTURE AND OF THE THERMOPLASTIC ALLOYS SO OBTAINED			
Thermoplastic alloy		1	2
40 45 Reactive mixture composition (parts by weight)	Polypropylene (PP) (Moplen® FL X020)	64.2	49.1
	t.butylperoxypivalate (in solution at 15%)	1.0	1.2
	Impact polystyrene (HIPS)	--	34.6
	Styrene	34.5	14.8
	Irganox® 1010 (*)	0.2	0.2
	SHT (**)	0.1	0.1
50 Alloy composition (parts by weight)	Polypropylene	60	45
	Polystyrene (PS)	30	--
	HIPS + PS	--	45
	PP-g-PS graft copolymer + PP-g-S	10	--
	PP-g-HIPS graft copolymer + PP-g-S	--	10

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(*) a phenolic stabilizer sold by Ciba-Geigy (based on pentaerythrityl tetrakis (3,5-ditert-butyl-4-hydroxyphenyl propionate)

(**) synthetic hydrotalcite

The alloys were prepared under dry conditions by using a turbomixer operating in a nitrogen atmosphere.

To the polypropylene in flakes and to the HIPS in pellets, if any, the peroxide and subsequently, under heating and in small batches, the styrene were added.

5 After the styrene was fully added in 1 hour, stirring was continued for additional 2 hours under slight cooling to prevent the temperature from exceeding 130 °C in order to avoid softening the consequent thickening of the polymer.

The products were gradually cooled and then stabilized with 0.2% of Irganox® 1010 and 0.1% of SHT, whereafter they were extruded at 210 °C.

10 Selective extractions with methylethylketone and chloroform and subsequent infrared analyses, carried out on the various fractions, revealed in both products the presence of about 10% of polystyrene-g-polypropylene graft copolymer. The maximum size of the dispersed styrene polymer phase was for at least 80% of less than 2 µm.

15 Table 2 shows the compositions and the main characteristics of the elastoplastic compositions prepared from the polymeric alloys defined in Table 1.

Table 2

ELASTIC AND RHEOLOGICAL CHARACTERISTICS OF THE ELASTOPLASTIC COMPOSITIONS				
Sample	1 (comparative)	2	3	
Ethylene-propylene-ethylidene-norbornene terpolymer * (Dutral® TER 537 E2)	64	64	64	
Polypropylene (Moplen® Q 30P)	6.5	--	--	
25 Impact- polystyrene	6.5	--	--	
Thermoplastic alloy 2 of Table 1	--	13	--	
Thermoplastic alloy 1 of Table 1	--	--	13	
Master FX-RA-148 of Schenectady **	7.2	7.2	7.2	
p-toluenesulfonic acid	0.4	0.4	0.4	
30 Zinc oxide	7.4	7.4	7.4	
Oil	8	8	8	
PP/(PP + EPDM) ratio	0.169	0.155	0.196	
Pressure, in kg/cm ² , (KPa) recorded in TR 15 at 230 °C and a take-off rate of 9.5 cc/minute	150 (15000)	110 (11000)	75 (7500)	
35 Extrudate appearance	not smooth	smooth	smooth	
Tension set at 23 °C at 200%, %	breaks	20	20	
Compression set at 100 °C, 22 hours, %	45	27	23	

40 * containing 50% by weight of extender oil.

** Master FX-RA-148 is composed of :

50% of phenolic resin SP 1045

50% of barium sulfate.

45 The compositions reported herein, analogously with all the ones of the following examples, were prepared by introducing the polymeric components into a Brabender internal mixer and, after a short mixing period, by adding the curing system and subsequently the zinc oxide and the extender oil.

Thereafter the mixing was carried out for 3 minutes at a temperature of 200 °C.

50 The composition was then discharged from the internal mixer and was subjected to the following determinations :

- Processability, by measuring the head pressure during extrusion tests in an extruder. In such tests, the extruder was of type TR 15 (single-screw, 15 mm diameter), and it was operated at a temperature of 230 °C, with a 2.5 mm ID die, a LD ratio = 20 and at a take-off rate of 9.5 cc/minute.

- Tension set at 200%, measured at 23 °C, according to ASTM D-412.

55 - Compression set after 22 hours at 100 °C, according to ASTM D-395.

The advantages deriving from the use of the alloys comprising the amorphous styrene phase are apparent also for the harder elastoplastic compositions having higher crystalline olefin resin/elastomeric terpolymer ratios (see Table 3).

Dutral® TER 537 E2 is an EPDM rubber produced by DUTRAL S.p.A.; Moplen® Q 30P is sold by HIMONT ITALIA S.p.A.

Table 3

ELASTIC AND RHEOLOGICAL CHARACTERISTICS OF ELASTOPLASTIC COMPOSITIONS BASED ON THERMOPLASTIC ALLOY 2				
Sample	1 (comparative)	2	3 (comparative)	4
Dutral® TER 537 E2 *	59	59	52	52
Moplen® Q 30P	18	--	25	--
Thermoplastic alloy 2 (see Table 1)	--	18	--	25
Master FX-RA-148 produced by Schenectady	7.2	7.2	7.2	7.2
p-toluenesulfonic acid	0.4	0.4	0.4	0.4
Zinc oxide	7.4	7.4	7.4	7.4
Oil	8	8	8	8
PP (PP + EPDM) ratio	0.379	0.215	0.490	0.302
Pressure, in Kg/cm ² (KPa) recorded in TR 15 at 230 °C at a take-off rate of 9.5 cc/minute	50 (5000)	25 (2500)	40 (4000)	25 (2500)
Tension set at 23 °C at 200%, %	breaks	breaks	breaks	breaks
Compression set at 100 °C, 22 h, %	36	34	64	55
Hardness, Shore A	70	70	85	85

* containing 50% by weight of extender oil.

In the elastoplastic compositions of Table 2 (samples 2 and 3) and Table 3 (samples 2 and 4) more than 90% of the particles of the dispersed phase have a maximum size below 2 μ m.

Example 2

Table 4 shows the compositions of various alloys utilized for preparing the elastoplastic compositions.

Table 4

COMPOSITIONS OF THE ALLOYS UTILIZED FOR PREPARING ELASTOPLASTIC COMPOSITIONS		
Alloy	1	2
Moplen® Q 30P	50	50
Ultrastyr® W 275 *	50	--
Ultrastyr® AES Y42 **	--	50

* Ultrastyr® W 275 is a polystyrene grafted with 10% of Dutral® TER 044 (Ultrastyr is a product sold by MONTEDIPE S.p.A.)

** Ultrastyr® AES Y42 is a styrene-acrylonitrile copolymer (75% S - 25% AN) grafted with 30% of Dutral® TER 044 (EPDM rubber sold by DUTRAL S.p.A.).

Table 5 shows compositions and main characteristics of the compositions prepared from the above-

indicated alloys. In alloy 1, the maximum size of more than 80% of the dispersed phase present in alloy 1 was of 40 μm .

Table 5

ELASTIC AND RHEOLOGICAL CHARACTERISTICS OF ELASTOPLASTIC COMPOSITIONS		
Sample	1	2
Dutral® TER 537 E2	64	64
Thermoplastic alloy 1 (see Table 4)	13	--
Thermoplastic alloy 2 (see Table 4)	--	13
Master FX-RA-148 of Schenectady	7.2	7.2
p-toluenesulfonic acid	0.4	0.4
Zinc oxide	7.4	7.4
Oil	8	8
PP/(PP + EPDM) ratio	0.169	0.169
Pressure, in Kg/cm ² (KPa) recorded in TR 15 at 230 °C at a take-off rate of 9.5 cc/minute	80 (8000)	110 (11000)
Extrudate appearance	smooth	smooth
Tension set at 23 °C at 200%, %	21	20
Compression set at 100 °C, 22 h, %	27	25

* containing 50% by weight of extender oil

Example 3

Table 6 shows the compositions of some alloys utilized for preparing elastoplastic compositions.

Table 6

COMPOSITION OF THE ALLOYS UTILIZED FOR PREPARING ELASTOPLASTIC COMPOSITIONS				
Alloy	1	2	3	4
Moplen® Q 30P	42	50	62	69
Kraton® G 1652	58	50	38	31

Table 7 shows the compositions and characteristics of the compositions prepared with such alloys.

In alloys 1-4, more than 90% of the particles of the dispersed (polystyrene) phase has a maximum size below 1-2 μm .

Table 7

ELASTIC AND RHEOLOGICAL CHARACTERISTICS OF COMPOSITIONS BASED ON THERMOPLASTIC ALLOYS OF						
TABLE 6						
Sample	1	2	3	4	5 comparison	6 comparison
Dutral® TER	64	64	64	64	70.8	66
537 E2 (50% oil)	--	--	--	--	6.2	11
Moplen® Q 30P	13	--	--	--	--	--
Alloy 1	--	13	--	--	--	--
Alloy 2	--	--	13	--	--	--
Alloy 3	--	--	--	13	--	--
Alloy 4	--	--	--	--	--	--
Master	7.2	7.2	7.2	7.2	7.2	7.2
FX-RA-148 of						
Schenectady	0.4	0.4	0.4	0.4	0.4	0.4
p-toluenesulfonic						
acid						
Zinc oxide	7.4	7.4	7.4	7.4	7.4	7.4
Oil	8	8	8	8	8	8
PP/(PP + EPDM) ratio	0.147	0.169	0.200	0.219	0.15	0.25
Pressure, in Kg/cm ² (KPa) recorded in TR	130 (13000)	115 (11500)	90 (9000)	70 (7000)	90 (9000)	55 (5500)
15 at 230 °C at a take-off rate of 9.5 cc/minute						
Extrudate appearance	smooth	smooth	smooth	smooth	melt fracture	melt fracture
Tension set at 23 °C at 200%, %	--	--	16	--	breaks	breaks
Compression set at 100 °C 22 h, %	23	22	24	25	16	24

Also these compositions prove to be very interesting as they exhibit excellent elastic characteristics also at high temperatures as well as a good processability.

5 Example 4

Example 4 regards compositions in which a non-halogenated phenolic resin alone is used as a curative. Table 8 shows the compositions and characteristics of such compositions.

Table 8

COMPOSITION AND CHARACTERISTICS OF ELASTOPLASTIC COMPOSITIONS CURED IN THE ABSENCE OF P-TOLUENESULFONIC ACID			
Sample	1	2	3 (comparison)
Dutral® TER 537 E2 (50% oil)	68	68	71.5
Moplen® Q 30P	8.5	8.5	9
Ultrastyr® W 275	4	--	--
Kraton G® 1652	--	4	--
Master FX-RA-148 of Schenectady	4	4	4
Zinc oxide	7.5	7.5	7.5
Oil	8	8	8
PP/(PP + EPDM) ratio	0.200	0.200	0.20
Pressure, in Kg/cm ² recorded in TR 15 at 230° C at a take-off rate of 9.5 cc/minute	100	100	80
Extrudate appearance	smooth	smooth	melt fracture breaks
Tension set at 23° C at 200%, %	14	14	
Compression set at 100° C, 22 h, %	16	18	20
Hardness, Shore A	56	50	58

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Claims

1. Elastoplastic compositions comprising a continuous crystalline polyolefin phase and at least two polymeric phases dispersed in the polyolefin phase, one of which consists of a dynamically cured EPDM rubber and the other of an amorphous thermoplastic styrene polymer, wherein the EPDM rubber is a copolymer of propylene with ethylene and/or with an alpha olefin of formula $\text{CH}_2=\text{CHR}$ in which R is an alkyl radical having 2-10 carbon atoms, and with a copolymerizable diene, the weight ratio between polyolefin resin and EPDM rubber ranges from about 10/90 to about 75/25 and the weight ratio between styrene polymer and polyolefin ranges from about 10/90 to about 60/40.
2. The compositions of claim 1 in which at least 80% of the particles of the two dispersed phases has a maximum size of 5 μm .
3. The compositions of claim 1 in which the EPDM rubber is fully cured.
4. The compositions of claim 1 and/or 2 in which the polyolefin is high isotactic polypropylene and the styrene polymer is selected from polystyrene, impact-resistant polystyrene and copolymers of styrene with acrylonitrile containing up to 30% by weight of acrylonitrile.
5. The compositions of claim 1 and/or 2 in which the styrene polymer is a styrene-ethylene-butene-styrene block copolymer.
6. The compositions of one or more of claims 1 to 3 in which the styrene polymer is compatibilized with the olefin polymer by using a graft copolymer of styrene on a polyolefin.
7. The compositions of one or more of claims 1 to 3 in which the compatibilizing agent is a graft copolymer of an EPDM rubber on polystyrene or on a copolymer of styrene with acrylonitrile.
8. The compositions of the preceding claims in which the compositions comprise an extender oil and a mineral filler.

9. A process for preparing the compositions as claimed in one or more of the preceding claims in which the olefin polymer and the styrene polymer, homogeneously mixed with each other, are added and mixed with the EPDM rubber and the curative system is added to the resulting homogeneous blend, mastication of the blend being continued at a temperature between about 150° and 280° C for a time period in which rubber curing occurs.

10. The process of claim 8 in which the EPDM rubber undergoes a dynamic cure in the presence of a curing system consisting of a non-halogenated phenolic resin utilized in an amount of from 1 to 10 parts by weight for 100 parts of rubber.

11. The process of claims 8 or 9 in which the EPDM rubber is fully cured.

12. Shaped articles obtained from the compositions defined in one or more of claims 1 to 7 and prepared according to a process as defined in one or more of claims 8 to 10.

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